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QUANTIFICATION OF THE EFFECT OF HYSTERESIS ON THE ADIABATIC TEMPERATURE CHANGE IN MAGNETOCALORIC MATERIALS

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ABSTRACT — We quantify the effect of hysteresis on the performance of the magnetocaloric first order material $Gd_5Si_2Ge_2$ undergoing an ideal active magnetic regenerator (AMR) cycle. The material is carefully characterized through magnetometry (VSM) and calorimetry (DSC) in order to enable an accurate model description of the phase transition at varying magnetic fields and temperatures. Using detailed experimental property data, a Preisach type model is used to describe the thermal hysteresis effects and simulate the material under realistic working conditions. We find that the adiabatic temperature change is limited by a significant fraction of the thermal hysteresis.

1. Introduction

Magnetocaloric first order materials have a coupled magnetic and structural transition, giving rise to both magnetic and thermal hysteresis in the characteristic material properties, i.e. magnetization and heat capacity. Hysteresis causes some degree of entropy production in each cycle, but more importantly also makes the material properties history dependent and thus non-single valued functions of both magnetic field and temperature. This makes determination of the adiabatic temperature change (ΔT_{ad}) non-trivial. In order to account for the material history dependence we employ a non-equilibrium Preisach-type model.

2. Methods

Experimental characterization of $Gd_5Si_2Ge_2$

The material used for this work is the first order magnetocaloric material $Gd_5Si_2Ge_2$. The sample is a solid rectangular piece of about 60 mg, provided by Ames Laboratory [1].

The data to be modelled was obtained through isothermal magnetization measurements. Each isotherm was obtained by the standard procedure of starting at the lowest temperature (230 K) and measuring magnetization and demagnetization curves with a maximum applied field of $\mu_0 H = 1.6$ T. The temperature is varied by chosen steps (1 K around the transition) and the procedure is repeated until the maximum temperature is reached (310 K). A selection of the measured isotherms is shown in Fig. 1 (symbols). Measurements were performed with a careful temperature control and a low field ramp rate of 0.05 T/min, in order to avoid temperature overshooting and the MCE affecting the measurements, which was unfortunately not quite achieved, as seen at $T = 272$ K.

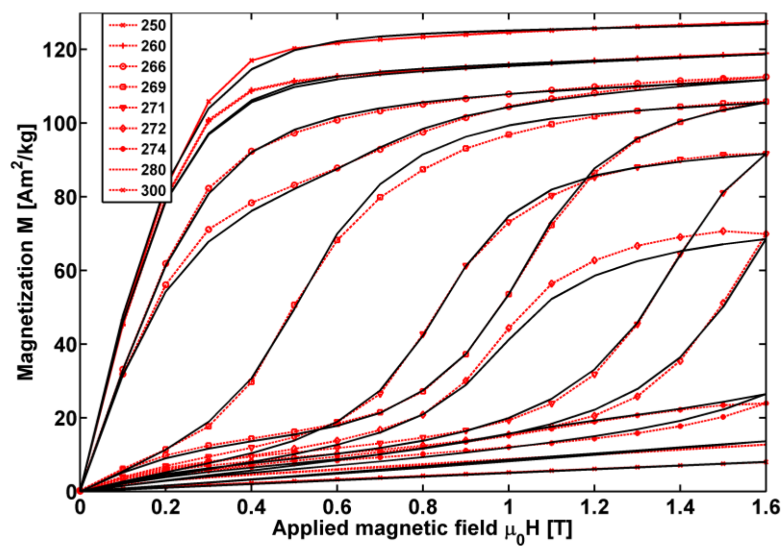

Fig. 1. Isothermal magnetization measurements on $Gd_5Si_2Ge_2$ (dashed symbols) and the corresponding Preisach model simulations (solid lines).

Furthermore, the sample was characterized by a zero field DSC scan from 240 K – 310 K to obtain a baseline for the heat capacity (and entropy) in the model. The heat capacity is approximately constant, $c_p \approx 300$ J/kg/K, away from the phase transition.

The Preisach model

A Preisach-type model is employed to describe the out-of-equilibrium aspects of the first order material. The Preisach approach to magnetic modelling utilizes a superposition of hysteretic bistable units, each characterized by two fields, switching the unit state between the purely ferromagnetic (FM) and paramagnetic (PM) phases. A detailed description of the applied Preisach model can be found in [1, 2]. In short, the magnetization M and entropy s are given as

$$M(H,T) = X \cdot M_{\mathrm{fm}}(H,T) + (1-X) \cdot M_{\mathrm{pm}}(H,T), \qquad s(H,T) = X \cdot s_{\mathrm{fm}}(H,T) + (1-X) \cdot s_{\mathrm{pm}}(H,T), \tag{1}$$

where the path dependent ferromagnetic phase fraction, X, is the main output of the Preisach model. The pure phase magnetization behavior, $M_{\mathrm{fm/pm}}$, is approximated from the non-hysteretic low and high temperature magnetization data (Fig. 1). The field independent part of the pure phase entropy, $s_{\mathrm{fm/pm}}$, is obtained from the DSC data and the magnetic contribution calculated from $M_{\mathrm{fm/pm}}$.

The Preisach model can now be used simulate the out-of-equilibrium transformations that the material undergoes during temperature and field changes. A simulation of the magnetization experiment is shown in Fig. 1 (solid lines). The hysteretic behavior is captured across the whole temperature range, including around the transition where the material only undergoes a partial phase transition.

3. Results

The model is applied to simulate an ideal AMR-cycle: adiabatic magnetization at T_i → isofield cooling back to T_i → adiabatic demagnetization at T_i → isofield heating back to T_i. The hysteretic path dependence is taken into account, but the entropy production is ignored.

An example of such a cycle is shown in Fig. 2(inset), where the path is denoted by the triangular markers. Starting at 267 K, the magnetization from 0 – 1.6 T results in ΔT_{ad} = 4.4 K. The enveloping entropy curves show the full transition from the pure FM to PM state and back, by scanning temperature, in 0 and 1.5 T applied field. It is seen that the s-T path is limited by the thermal hysteresis region, effectively reducing ΔT_{ad} by 1.5 K, compared to the completely reversible case (central dot-dashed lines). It is noted that the maximum hysteresis loss during a magnetization cycle is about 50 J/kg (maximum (M,H)-loop area in Fig. 1), resulting in an approximate temperature increase of 0.1 K during a half cycle, making it a small perturbation to ΔT_{ad}.

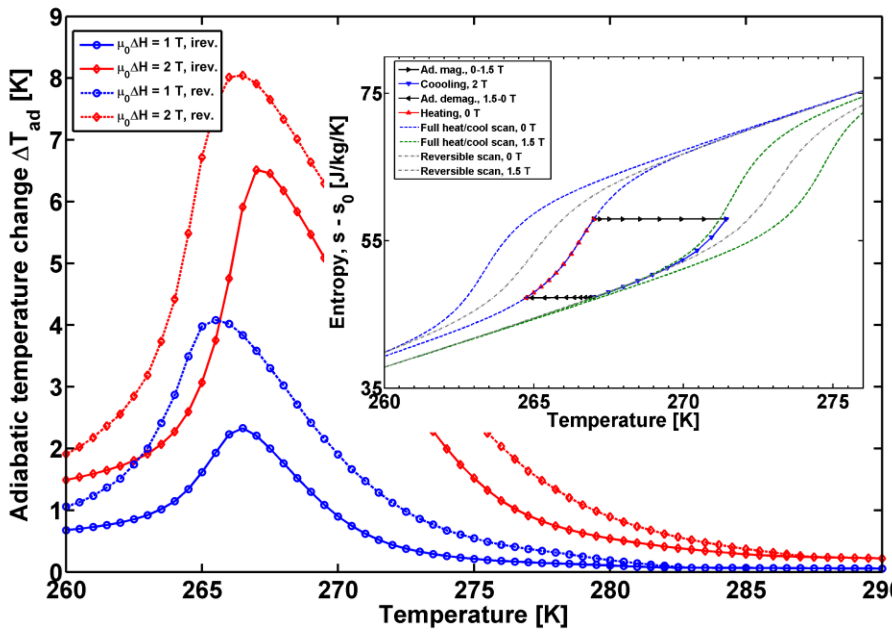

Fig. 2.(Main figure) The simulated adiabatic temperature change during magnetization at field changes of 1 T (circles) and 2 T (diamonds). Solid and dashed lines show the simulation of the irreversible and reversible cases, respectively. (Inset figure) A simulation of an AMR cycle at 267 K, with max field of 1.5 T. The dashed center lines show the corresponding reversible case.

A full $\Delta T_{ad}(T)$ experiment has been simulated, starting at low temperature and stepwise increasing the temperature, each time calculating ΔT_{ad} for an applied magnetic field. Two solid curves are shown in Fig. 2, corresponding to applied maximum fields of 1 T and 2 T. The dashed curves represent the completely reversible case.

Conclusion

These results show that if the magnetic field cannot force a complete PM-to-FM transition, then ΔT_{ad} is effectively limited by a significant fraction of the thermal hysteresis around the transition temperature.

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